

## ULTRAFILTRATION DE-ASHING PROCESS FOR SOLVENT REFINED COAL

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### INTRODUCTION

Most ultrafiltration (UF) studies have focused on aqueous systems where solutions are concentrated by the removal of water<sup>(1)</sup>. References to the UF of non-aqueous solutions are infrequent. The paucity of studies on non-aqueous solutions has been due primarily to three reasons. First, present-day membrane technology is primarily directed toward the demineralization of saline and brackish waters. Second, the organic fouling in water of conventional cellulosic type membranes has limited their use in non-aqueous, organic solutions. Third, the low price of energy during the last two decades did not encourage engineers to develop low pressure, thus low energy techniques of separation which were capital intensive. But now with the necessity for energy conservation and the development of membranes made of new polymeric materials, ones which provide both good mechanical characteristics and good resistance against chemical attack, membrane technologies are being applied to non-aqueous systems<sup>(2,3)</sup>.

Virtually all of the coal liquefaction processes require the separation of ash particles from the coal derived liquids in order to qualify the product as a furnace fuel. The SRC, Solvent Refined Coal, process produces a liquid having about 5% ash; a reduction of ash to below 0.1% is necessary to obviate the use of electrostatic precipitators in the furnace, thus reducing capital costs in product utilization. A further de-ashing to less than 0.005% would qualify SRC as a gas turbine fuel. Also, since most of the sulfur in SRC (0.55%) is concentrated in the ash; a UF de-ashing process would partially obviate the need for extensive desulfurization of the product<sup>(4)</sup>. A typical analysis is shown in Table 1. The asphaltenes (benzene soluble, pentane soluble) are believed to be responsible for the high viscosity of the product. A separation of these asphaltenes from the fuel could serve to avert viscosity-related problems in use. These asphaltenes could be further treated by thermal hydrocracking to produce more oil.

The objective in this investigation was to study the de-ashing (also de-sulfurization) of SRC using UF membranes including ones of fixed-charge, sulfonic acid nature. Principally such parameters as feed viscosity, temperature, use of solvents, cross-flow velocity, nature of the membrane and its fouling were of interest.

Table I

Analysis of SRC (SAMPLE from Filter feed from INDIANA V Coal)

Specific gravity at 90°C	1.087
Viscosity at 105°C	19.8 (cp)
Oils (Pentane solubles)	65.0% (wt. %)
Asphaltenes (Pentane Insolubles)	27.9% (wt. %)
Organic Benzene Insolubles	5.7% (wt. %)
Ash (Inorganic Insolubles)	5.45% (wt. %)
Sulfur	0.55% (wt. %)

EXPERIMENTAL

A. Apparatus: The system used in this investigation is shown in Fig. 1. The 2.0 l stainless steel reservoir was heated by heating tapes and its temperature controlled by a variac. The device readily withstands pressures up to 1500 psi. The operating pressures were provided by compressed nitrogen gas applied directly to the reservoir. The flow lines were made of 0.5 in. schedule - 40 pipes. Re-circulation rates were monitored with a high temperature and pressure (up to 1400 psig. and 205°C) dial indicator purgometer Model 10A2227, manufactured by Fischer and Porter Co. of Warminster, Pennsylvania. The operating temperature was measured at the exit of the cell with a stainless steel Weston thermometer. A special positive displacement pump from the Viking Pump Division, Model GGI950 with steel external direct drive unit, acted as a feed carrier to the system. This pump can handle many types of liquids with viscosities up to 15,000 S.S.U. at temperatures up to 107°C.

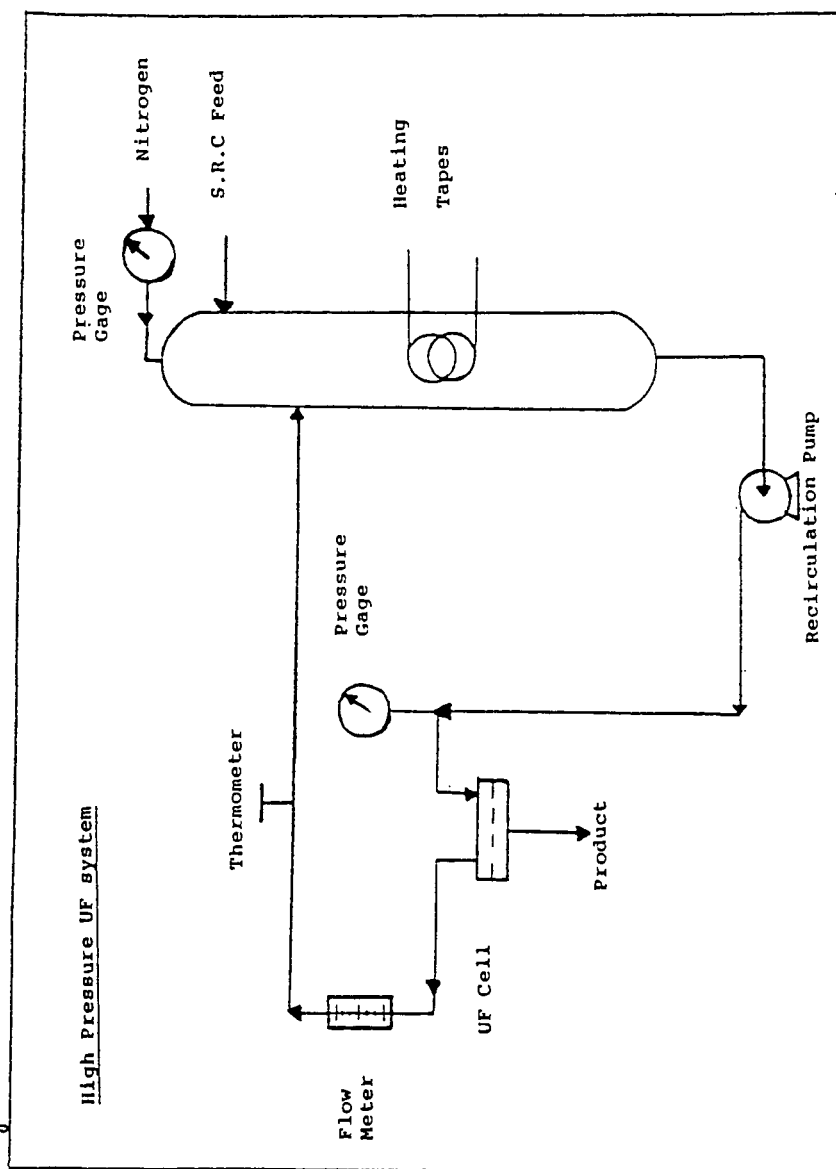
B. Membranes and their Conditioning: UF membranes are usually stored and conditioned in water before use. In this study, the first treatment was to condition the membranes in a non-aqueous environment. To avoid osmotic shock this transfer was accomplished gradually at regular intervals, every 10 minutes, by increasing the concentration of the solvent with respect to water, using a water miscible solvent such as methanol. Then the membrane could be transferred directly to any other non-aqueous solvent (toluene, hexane, kerosene, ...).

The membranes used in this study were cast from a solution of polyacrylamide onto a spun bonded polyester support. The membranes pore radii were about 1.5 nm and 70% pore volume (Poiseuille).

C. 24 CB - Creosote Solvent: This solvent was obtained from Allied Chemical, Semet Solvey Division of Morristown, New Jersey. This is a specially refined creosote distillate of tar obtained by the high temperature carbonization of bituminous coal, and further processed to remove excess crystalline salts and make the oil more fluid. The 24-CB creosote oil has a specific gravity at 38°C/15.5°C of 1.06, and its viscosity is about 2.1 Cp at 100°C.

D. Analysis for Ash: A muffle furnace operating at temperatures up to 950°C was used for the analysis for inorganic ash. Weighted samples of the feeds and the permeates of SRC (each weighing about 10 g) were dried overnight at about 100°C; following this the temperature of the furnace was increased by 50°C increments every half hour until 900°C and then kept there for 2-3 hours, after which the red-dish brown powder which resulted was cooled in a dessicator and

Figure 1.



weighted. To insure total decarbonation, the process at 900°C was continued until no further weight loss was observed.

## RESULTS AND DISCUSSION

As shown in Figure 2, an increase in temperature from 85°C to 100°C increased the flux by a factor of about 2.5. It is known that high temperatures reduce the viscosity of the fluid. With SRC, on going from 85°C to 100°C the viscosity was lowered from about 42.0 Cp to about 20.8 Cp, or by a factor of 2.0. As for ash rejection (99.8% at 85°C; 99.9% at 100°C) this improves with higher temperatures and with higher axial velocity of the fluid due to its lowered viscosity. From Figure 3, one can see that high pressures tend to increase the resistance to flow because under high pressures particles are forced against the membrane and possibly into the pores. These effects create a polarized boundary layer which may prevent higher fluxes. We observed that at the beginning of the run, at high pressures ( $15.51 \times 10^5$  Pascals or 225 psig) the rejection is low compared to the one measured at the end of the run<sup>(5)</sup>. High pressures force the smaller particles into the larger pores, which are responsible for the low rejection. After the gel slime has formed, the rejection is usually 100% but the fluxes are much lower.

UF of SRC was run, at the same temperature and pressure but at different axial velocities, as shown on Figure 4. At high axial velocities the fluxes leveled off smoothly to reach a higher steady state level; at low velocities, fluxes decreased sharply as shown by the curve for the velocity of  $v = 305$  cm/sec. Higher velocities apparently prevented the formation of the gel slime on the surface of the membrane, and reduced the thickness of the boundary layer by promoting turbulence, in the flow of the fluid in the cell. As for ash, rejection, (99.90%<sup>1</sup>, 99.89%<sup>2</sup>, 99.80%<sup>3</sup>) it increases with increasing velocities for the same reasons given above.

The permeabilities of diluted SRC with 24-CB Creosote oil is shown in Figure 5 (Curve #2). It shows that a 10% dilution almost doubled the fluxes. This again is explained by the fact that Creosote oil has a low viscosity (about 2.0 Cp at 94°C) compared with the SRC viscosity of 35.8 Cp at 94°C, this reduces the viscosity of the SRC solution and makes the fluid much easier to flow. Ash rejection also increased with dilution, 100% compared with that of pure SRC (99.9%). One observes that dilution has the same effect as temperature; both serve to reduce the SRC viscosity.

With toluene, as shown in Figure 5 (Curves #3 and 4), one can see a strong effect of dilution. A 10% dilution increased the flux by a factor of 3.5; a 25% dilution increased the flux by a factor of 6.5. Ash rejection also increased with dilution, 99.99 - 100% compared with 99.9% for undiluted SRC.

From the results shown in Figure 5, one observes that toluene served much better than 24-CB Creosote oil, primarily because it has a lower viscosity than creosote oil.

- 1 at  $v = 366$  cm/sec.  
2 at  $v = 335$  cm/sec.  
3 at  $v = 305$  cm/sec.

## ACKNOWLEDGEMENTS

The author wishes to acknowledge the generous support of Sonatrach, Co. in Houston, Texas and Professor Harry P. Gregor for his supervision and advice.

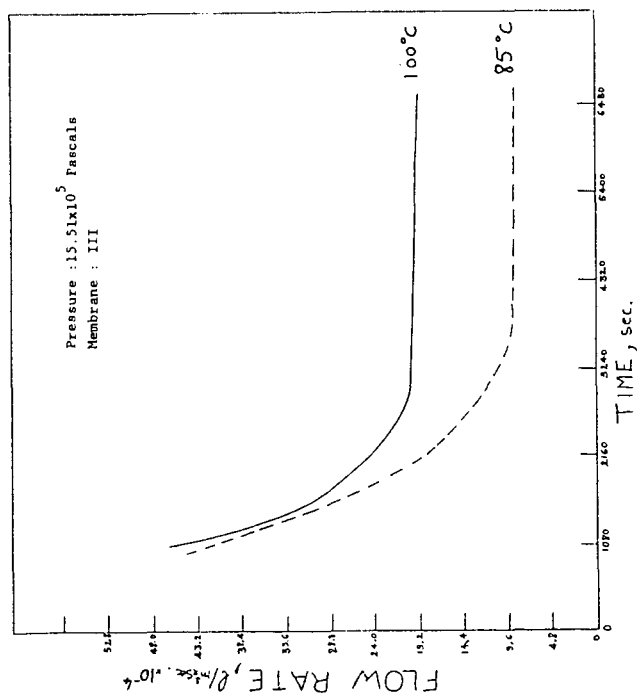


Figure 2. Pure SRC Fluxes as a Function of Temperature.

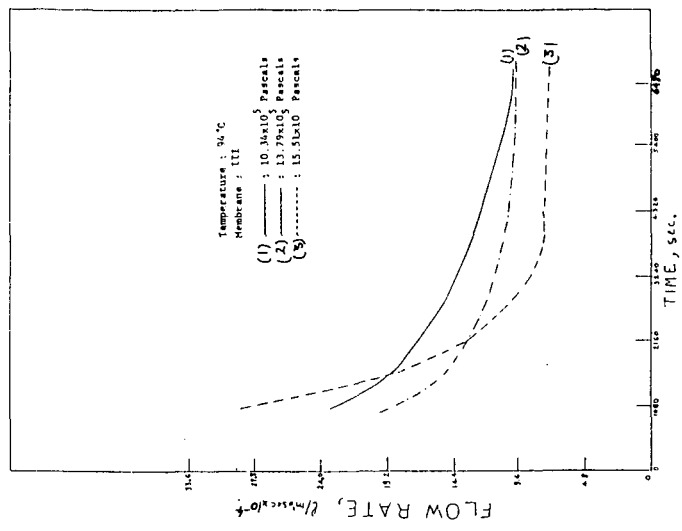


Figure 3. Pure SRC Fluxes as a Function of Pressure.

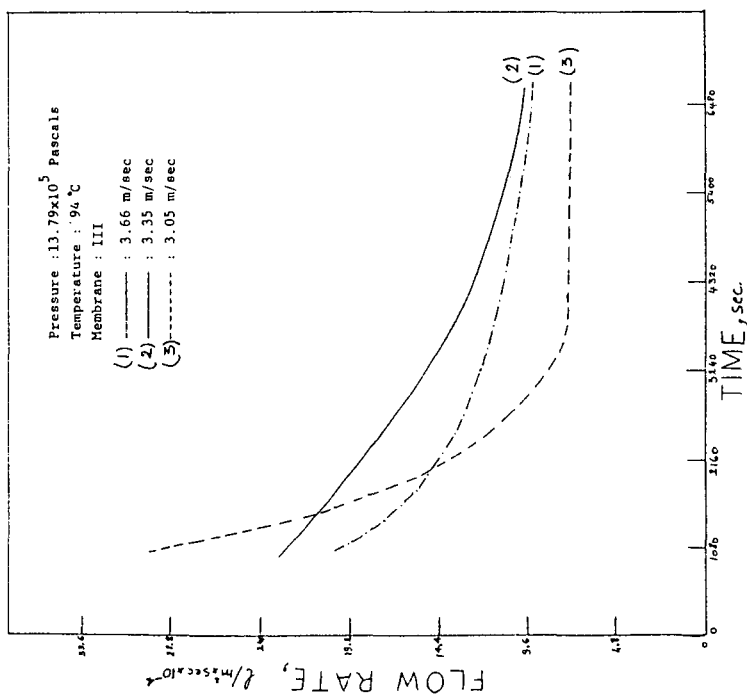


Figure 4. Pure SRC Fluxes as a Function of Axial Velocity.

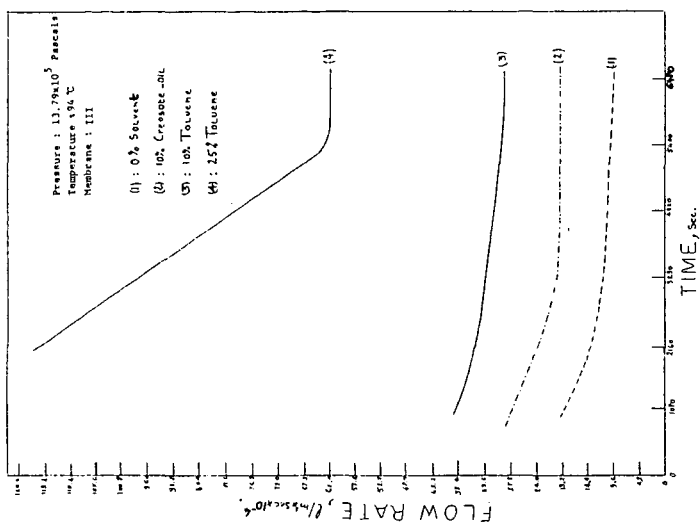


Figure 5. Comparison of Fluxes of SRC Diluted with Cresote and Toluene.

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